REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. AGENCY USE ONLY (Leave blank)

2. REPORT DATE 20 December 1989

3. REPORT TYPE AND DATES COVERED Final Technical Report: 11/15/88-10/15/89

4. TITLE AND SUBTITLE
Pulsed Laser Deposition of High T_c Superconducting Thin Films

5. FUNDING NUMBERS
(2)
2306/C1

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8. PERFORMING ORGANIZATION REPORT NUMBER
AFOSR-TR- 89-1875

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
USAF, AFSC
Air Force Office of Scientific Research
Building 410
Bolling AFB, DC 20332-6448

10. SPONSORING/MONITORING AGENCY REPORT NUMBER
F49620-89-C-0017

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited.

12b. DISTRIBUTION CODE

13. ABSTRACT (Maximum 200 words)

Superconducting thin films have been deposited *in-situ* on several substrate materials using pulsed excimer laser deposition. On the standard oxide substrates, excellent films were obtained. They had high transition temperatures with narrow transition widths, metallic conductivity in the normal state, low room-temperature resistivity, high critical currents, c-axis orientation, and epitaxial alignment with the substrate. On the more technologically relevant substrates of sapphire and silicon, world record, though less optimal, results were obtained. The transition temperatures were high and metallic conductivity was obtained in the normal state. However, the room-temperature and microwave surface resistivities were higher and the critical currents lower than for the above substrates. These diminished transport properties correlate with the imperfect alignment and epitaxy of the superconductor substrate. For silicon substrates, a buffer layer is required due to high reactivity even at low temperatures. The best results were obtained on clean, hydrogen-terminated surfaces rather than oxidized silicon. Epitaxial alignment was achieved, but there was a substantial spread in orientations, accounting for the diminished transport properties.

14. SUBJECT TERMS

15. NUMBER OF PAGES 5

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
Unclassified	Unclassified	Unclassified	UL

NSN 7540-01-280-5500

Standard Form 298 (Rev. 2-89)

AD-A216 507

90 01 04 141

Final Technical Report on AFOSR Contract F49620-89-C-0017

Title: Pulsed Laser Deposition of High T_c Superconducting Thin Films

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Statement of Work: We will use laser deposition to grow thin film superconductors that cover a wide range of compositions, substrates, and deposition conditions. These films will be characterized and provided for in-depth study and device processing both by ourselves and collaborators at Xerox and Stanford University and their associates.

Status of the Research Effort

During the last year of support on this contract, a new PolyGun laser deposition system was built and has been used to deposit excellent *in-situ* YBCO films on $SrTiO_3$, MgO, $LaAlO_3$, and yttrium-stabilized zirconia (YSZ), as well as, on the reactive substrates, Al_2O_3 and Si. The PolyGun system holds ten hot-pressed targets, mounted on the faces of a rotating polygon. The pulses from a 308 nm XeCl excimer laser are synchronized with the target wheel rotation to ablate the target or targets of interest during each revolution. For YBCO on sapphire or the standard substrates, this corresponds to hitting only one or more bulk YBCO targets per revolution. While the target wheel rotation is not necessary for a single-target case, the rotation does significantly reduce the target temperature rise by limiting its exposure to radiation from the hot substrate. The deposited films have better surface morphology as a consequence. Also the target wheel is surrounded by a water-cooled can with a small opening to allow for the laser ablation. This further reduces the heating of the targets and minimizes back-sputtering of one target onto another. The full advantage of the rotating polygon comes into play for multilayer film depositions and for the mixed deposition of film materials from more than one target. We have used this capability for YBCO on Si where buffer layers were deposited, as described below.

All of our YBCO laser depositions are carried out at an elevated substrate temperature in an oxygen atmosphere. The parameters are adjusted for *in-situ* growth; no post anneals of the films are performed. Use of a 2.45 GHz atomic oxygen

source does not appear to substantially improve the YBCO film quality at the optimum oxygen pressure and substrate temperature.

Films with sharp superconducting transitions ($\Delta T_c \approx 0.3K$) have been prepared by laser deposition on $SrTiO_3$, MgO, $LaAlO_3$, and YSZ. The electrical results we obtain are comparable to the best results reported in the literature to date. Since these substrates have the advantage of being relatively unreactive with YBCO at its growth temperature (about 700°C) and also allow for a high degree of epitaxy, films with good electrical properties are readily obtained. However, they also have several undesirable properties for electronic applications, namely, high dielectric constants, low mechanical strengths, and high costs. As a result, efforts have been made to deposit YBCO on substrates such as Al_2O_3 and Si which do not have the above disadvantages and, therefore, will be required for electronic devices. The difficulties in achieving this goal are that both Si and Al_2O_3 react with the superconductor during the high-temperature growth and annealing required to form the superconducting phase. Si, in addition to being more reactive than Al_2O_3, has the added problem that it rapidly forms a thin, poor quality surface oxide thereby preventing epitaxy.

For YBCO/Al_2O_3 good films are obtained only in a narrow substrate-temperature window of 650–670°C. For substrate temperatures too high, reaction occurs and the resulting films are transparent and insulating. For substrate temperatures too low, poor epitaxy and film orientation occur, resulting in lowered critical current densities. The resulting films have a high zero-resistance temperature, T_{c0}, of 86–88K and metallic conductivity in the normal state. The surface impedance at 13 GHz and 4.2K is 1 mΩ, only 20 times larger than for the best reported values for YBCO/$LaAlO_3$. In addition the critical current density, as determined from the magnetization hysteresis loop, is 5.4×10^6 A/cm^2 at 4.2K and 3kG, only 6 times lower than that for YBCO/$SrTiO_3$ and YBCO/$LaAlO_3$. These electrical parameters for YBCO/Al_2O_3 are the best reported to date. They are somewhat poorer than films on $SrTiO_3$, and the reasons can be found in the structural results.

For deposition on silicon substrates, three configurations were studied: (1) YBCO/SiO_2/Si, (2) YBCO/Si, and (3) YBCO/YSZ/Si. In case (1) the SiO_2 is a 150Å-thick, MOS-grade thermal oxide. The clean Si surfaces used for (2) and (3) are spin-etched free of oxide and hydrogen-terminated in a nitrogen-purged hood, then immediately introduced into the deposition chamber through a load lock. Their

surfaces have been demonstrated to be atomically clean and devoid of oxide prior to the start of a deposition, conditions that are usually necessary for successful epitaxial growth. For (1) and (2), depositions at several substrate temperatures from 550 to 700°C were performed. It was found that, except for very thick films (~1 μm), a buffer layer is required due to high reactivity even at substrate temperatures as low as 550°C. YSZ provides a good buffer, and our best results were obtained on clean, hydrogen-terminated surfaces rather than oxidized Si. For this case, (3), one could use a single YSZ target, but we used ZrO_2 and Y_2O_3 targets to study the influence of the amount of Y, and thereby the structure of this buffer layer, on the YBCO film. For the YSZ buffer, $(ZrO_2)_{1-x}(Y_2O_3)_x$ is deposited by adjusting the ratio of the number of pulses directed at the ZrO_2 and Y_2O_3 targets. Atomic-scale intermixing of the YSZ is assured, since each pulse only deposits about 0.2 Å of either oxide. The amount of Y_2O_3 in ZrO_2 was varied, and the best films were obtained with x near 0.1 where $(ZrO_2)_{1-x}(Y_2O_3)_x$ is cubic. The electrical results are reasonably good, with $T_{c0} = 86K$ and metallic conductivity in the normal state. However, T_{c0} is 5K lower and ρ(300K) 20 times larger than for our films on $SrTiO_3$. Epitaxial alignment of the YBCO with the Si was achieved, but there was a substantial spread in orientations, accounting for the diminished transport properties.

We, therefore, have demonstrated that the PolyGun laser-deposition technique can prepare thin films at relatively low temperatures without the need for high-temperature anneals. Also we have shown that multilayer films of particular compositions, e.g., YBCO/YSZ/Si, can all be fabricated with one computer-controlled set-up. This flexibility is not offered by any competing technique.

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G. A. N. Connell, D. B. Fenner, D. K. Fork, J. B. Boyce, F. A. Ponce, F. Bridges and T. H. Geballe, "YBCO Films and Buffer Layers Grown *In-Situ* by Pulsed Laser Deposition", Materials Research Society Symposium Proceedings on *High-Temperature Superconductors: Fundamental Properties and Novel Materials Processing,* Nov. 1989, in press.

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